

# **Reactivity-Worth Estimates of the OSMOSE Samples in the MINERVE Reactor R1-MOX, R2-UO<sub>2</sub> and MORGANE/R Configurations**

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**Nuclear Engineering Division**

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# **Reactivity-Worth Estimates of the OSMOSE Samples in the MINERVE Reactor R1-MOX, R2-UO<sub>2</sub> and MORGANE/R Configurations**

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by  
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## Abstract

An initial series of calculations of the reactivity-worth of the OSMOSE samples in the MINERVE reactor with the R2-UO<sub>2</sub> and MORGANE/R core configuration were completed. The calculation model was generated using the lattice physics code DRAGON. In addition, an initial comparison of calculated values to experimental measurements was performed based on preliminary results for the R1-MOX configuration.

## Introduction

The OSMOSE program (Oscillation in Minerve of isotopes in “Eupraxis” spectra) is a collaboration between the U.S. Department of Energy (DOE) and the Commissariat à l’Energie Atomique (CEA).(ref.1) It aims at measuring integral absorption rates of minor actinides by the oscillation technique in the MINERVE experimental facility located at the CEA Cadarache Research Center. The OSMOSE program also includes a complete analytical program to understand and resolve potential discrepancies between calculated and measured values. The OSMOSE program began in 2001 and will continue until 2009.

The Argonne National Laboratory has developed Monte Carlo and deterministic calculation models of the MINERVE facility to determine core and safety parameters and the reactivity worth of oscillated samples. Oscillation samples include calibration samples with different uranium enrichments and boron concentrations and the OSMOSE samples - separated actinides including <sup>232</sup>Th, <sup>233</sup>U, <sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>238</sup>U, <sup>237</sup>Np, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>242</sup>Pu, <sup>241</sup>Am, <sup>243</sup>Am, <sup>244</sup>Cm and <sup>245</sup>Cm. For the OSMOSE program, four different neutron spectra will be created in the MINERVE facility: an over-moderated UO<sub>2</sub> matrix (representative of a fuel processing plant or flooded storage cask), a UO<sub>2</sub> matrix in water (representative of LWRs), a mixed oxide fuel matrix (representative of cores containing MOX fuels), and an epithermal spectra (representative of under-moderated reactors). The different spectra are achieved by changing the experimental lattice within the MINERVE reactor.

The currently investigated core configurations, referred to as R2-UO<sub>2</sub>, R1-MOX and MORGANE/R, are representative of an over moderated LWR, an LWR loaded with MOX fuel, and an under-moderated LWR, respectively. The goal of this report is to report on the results of the reactivity worth of the OSMOSE samples in these configurations. The model of the samples is based on the specifications for fabrication of the samples. The analyses of the samples with as-built geometry and compositions will be performed after the samples have been chemically analyzed.

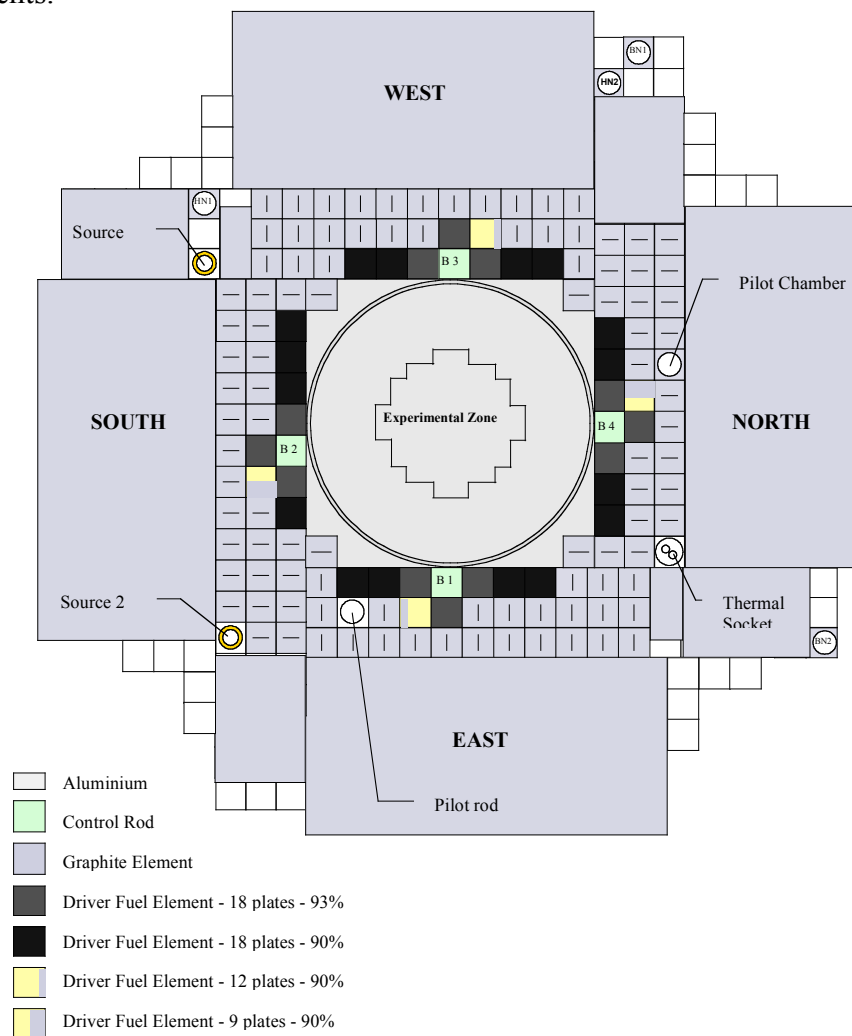
## Reactor Description

MINERVE is a zero power pool type experimental facility that is primarily used for neutronic studies. Built in the Nuclear Research Center of Fontenay-aux-Roses in 1959, it was transferred to the Nuclear Research Center (CEA) of Cadarache in 1977.

The maximum power of MINERVE is less than 100 W. Fuel elements are submerged under 3 meters of water. The driver fuel elements – made of highly enriched uranium plates – and the graphite reflector elements are mounted on 4 mobile grids to form the driver zone. In the center of the driver zone, an adjustable square cavity contains the experimental zone (ref.2). Figure 1 shows the layout of the MINERVE reactor with the R1-UO<sub>2</sub> experimental lattice.

The experimental zone is sub-critical and can be varied from one experiment to another. It is coupled to the driver zone and measurements are generally conducted in the center of the experimental zone.

The components of the control system – rods, automatic pilot rod, detectors – are located in the driver zone to avoid perturbations in the experimental zone. The automatic pilot rod maintains the power level in order to measure the reactivity change during oscillation measurements.



**Figure 1:** MINERVE Reactor

## Calculation Model

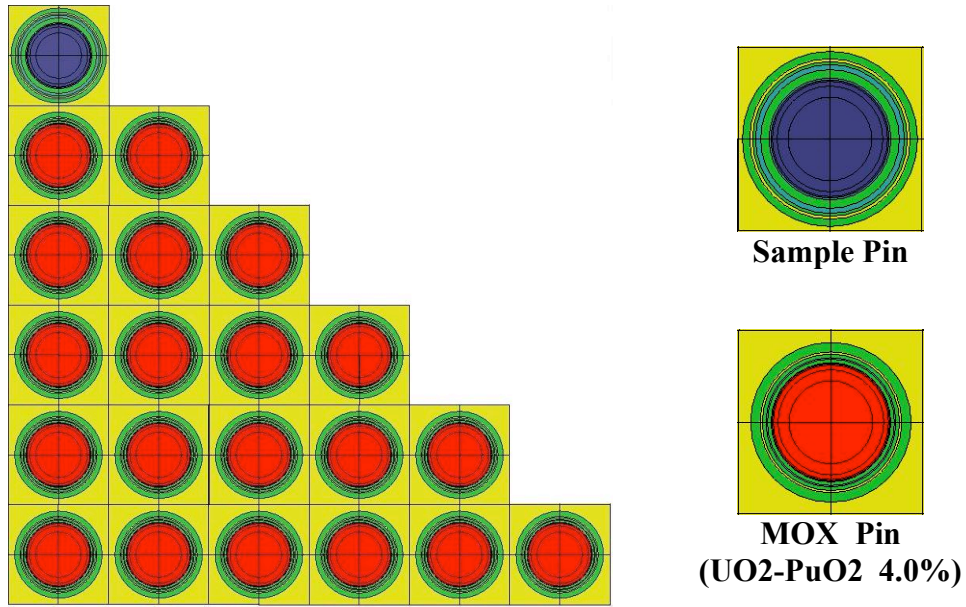
The DRAGON deterministic transport code (ref. 3) is used for the reactivity worth calculations. DRAGON is a lattice physics code based on the collision probability method. The two main components of the DRAGON code are a multi-group flux solver module and a one group collision probability (CP) tracking module. The different CP tracking modules perform the same tasks but with different levels of approximation.

The JPM tracking option uses the interface current technique at the level of each homogeneous zone associated with a specific geometry ( $J_{\pm}$  method). With this option, one can either build the complete collision probabilities matrix or generate a response matrix both of which can be processed by the general multi-group solver. The last method permits a non-iterative calculation of the one group neutron flux to be carried out using sparse matrix algebra.

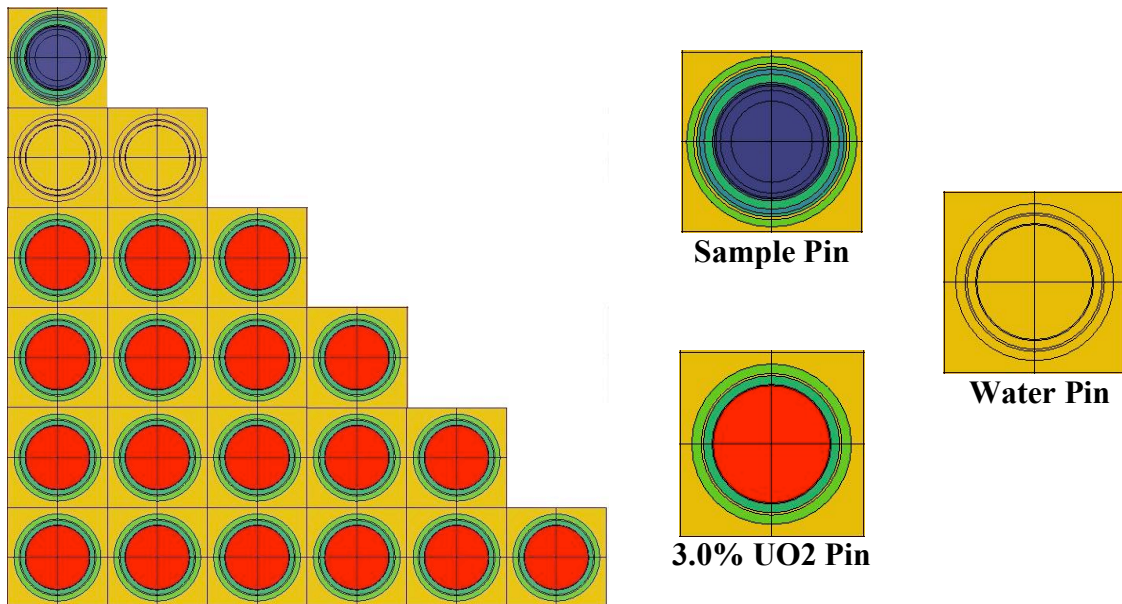
The SYBIL tracking option emulates the main flux calculation option available in the APOLLO-1 code, and includes a new version of the EURYDICE-2 code which performs reactor assembly calculations in both rectangular and hexagonal geometries using the interface current method. SYBIL is slightly more accurate than JPM due to the fact that it performs a complete calculation of the collision probabilities on the whole or a large part of the domain therefore avoiding a large number of interfaces for the angular flux approximation.

The EXCELL tracking option is used to generate the collision probability matrices for the cases having cluster, two dimensional or three dimensional mixed rectangular and cylindrical geometries. A cyclic tracking option is also available for treating specular boundary conditions in two dimensional rectangular geometries. After the collision probability or response matrices associated with a given cell have been generated, the multi-group solution module can be activated. This module uses the power iteration method and requires a number of iteration types. The thermal iterations are carried out by DRAGON so as to rebalance the flux distribution only in cases where neutrons undergo upscattering. The power iterations are performed by DRAGON to solve the fixed source or eigenvalue problem in the cases where a multiplicative medium is analyzed. The effective multiplication factor ( $k_{eff}$ ) is obtained during the power iterations. A search for the critical buckling may be superimposed upon the power iterations so as to force the multiplication factor to take on a fixed value.

The calculation model for the R1-MOX configuration consists of a two-dimensional ( $11 \times 11$ ) multi-cell mini-lattice corresponding to the experimental zone of the MINERVE reactor. Due to the symmetry of geometry, actually only  $1/8^{\text{th}}$  model is introduced. The model for the R1-MOX configuration is shown in Figure 2. The sample pin is located in the center, surrounded with MOX pins. The model for R2-UO2 is shown in Figure 3. It is similar to the R1-UO2 model (ref. 4), the only difference is that 8 water pins are surrounding the sample pin.



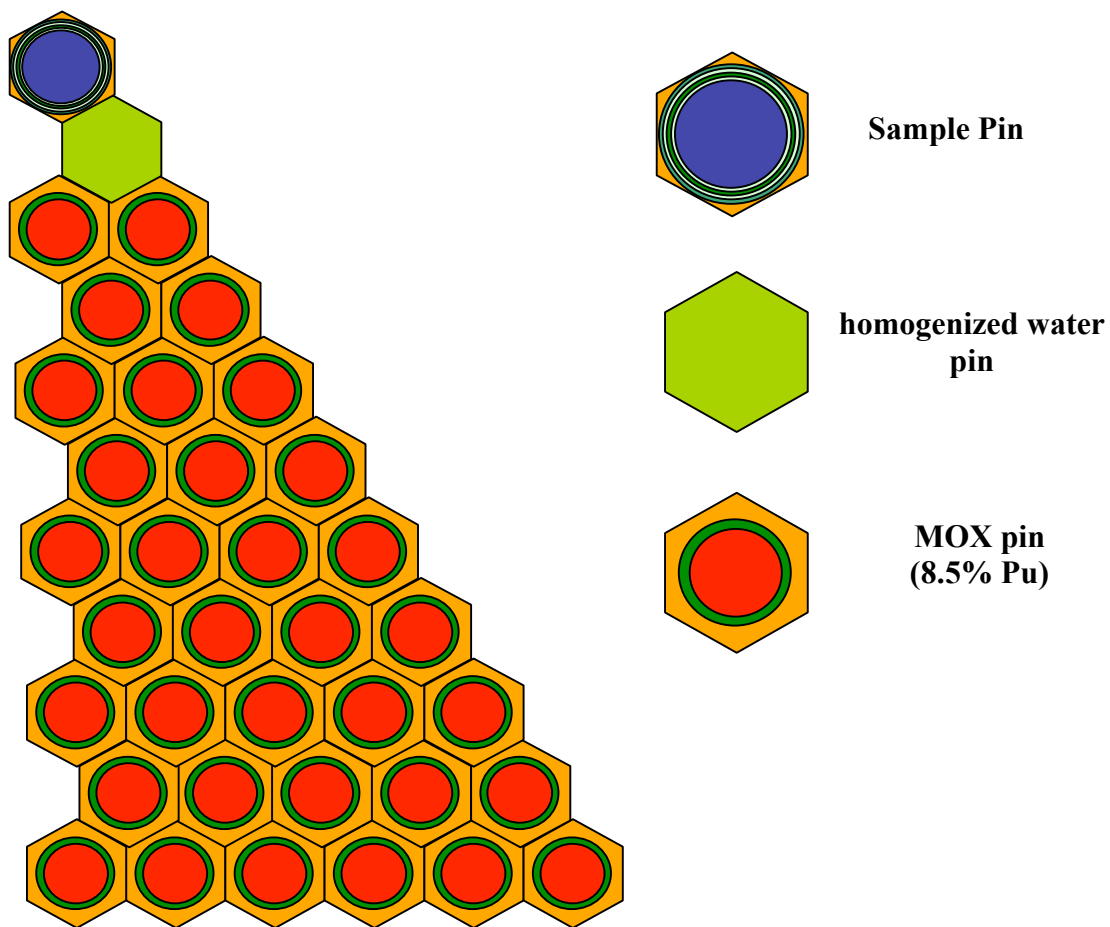
**Figure 2: DRAGON calculation model for R1-MOX configuration**



**Figure 3: DRAGON calculation model for R2-UO2 configuration**

For the MORGANE/R configuration, the hexagonal geometry capability of DRAGON is utilized, and the whole domain is composed of hexagonal cells. Due to the geometry limit of DRAGON, an approximate 1/12<sup>th</sup> model is developed for MORGANE/R, which is shown in Figure 4. Because the size of the sample pin cell is larger than the hexagonal lattice used for pin cell of normal MOX fuel, only the fuel and clad of the sample pin is kept and the guide tube is homogenized into the adjacent 6 hexagonal cells.

ENDF/B-VI and JEFF3.1 based WIMS-D4 format 172-group neutron libraries are used for the DRAGON calculation. The surface net current coupling option, SYBLIT, is used for the calculation.

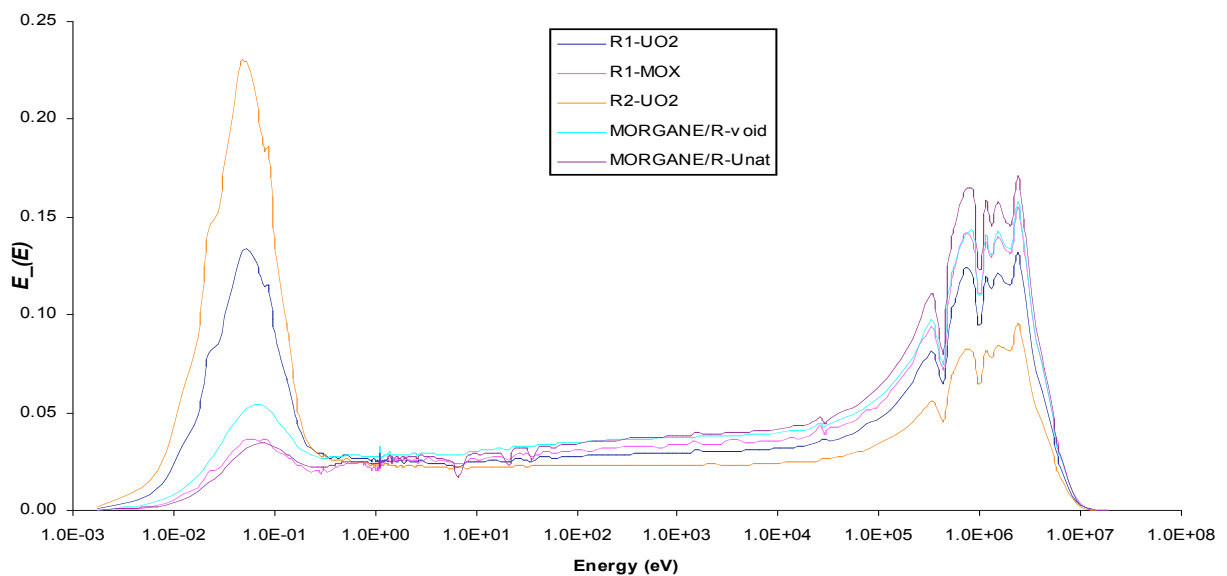


**Figure 4: DRAGON calculation model for MORGANE/R configuration**



## Spectra Comparison

A series of calculations were performed to calculate the spectra of OSMOSE configurations using the DRAGON code with the ENDF6-172 group library. The comparison of the spectra for different OSMOSE configurations are shown in Figure 5, and is consistent with expected results. The R2-UO<sub>2</sub> configuration displays the largest thermal flux, whereas, the MORGANE/R configuration, displays the smallest thermal fraction. This is expected since the R2-UO<sub>2</sub> configuration represents over-moderation and the MORGANE/R configuration is under-moderated.



**Figure 5: Comparison of spectra with different OSMOSE configurations**

## Reactivity-Worth Comparison

DRAGON calculations were also performed to obtain reactivity-worth estimates of the OSMOSE samples in the R2-UO<sub>2</sub>, R1-MOX and MORGANE/R configurations. The results are shown in Table 1, 2, 3 and Figures 6, 7, and 8.

The reactivity-worth estimate for a sample is not directly computed. The reactivity-worth of a sample is computed by subtracting the calculated reactivity of the configuration with the natural uranium sample in the experimental region from the calculated reactivity of the configuration with the sample in place. So for a given configuration, the reactivity-

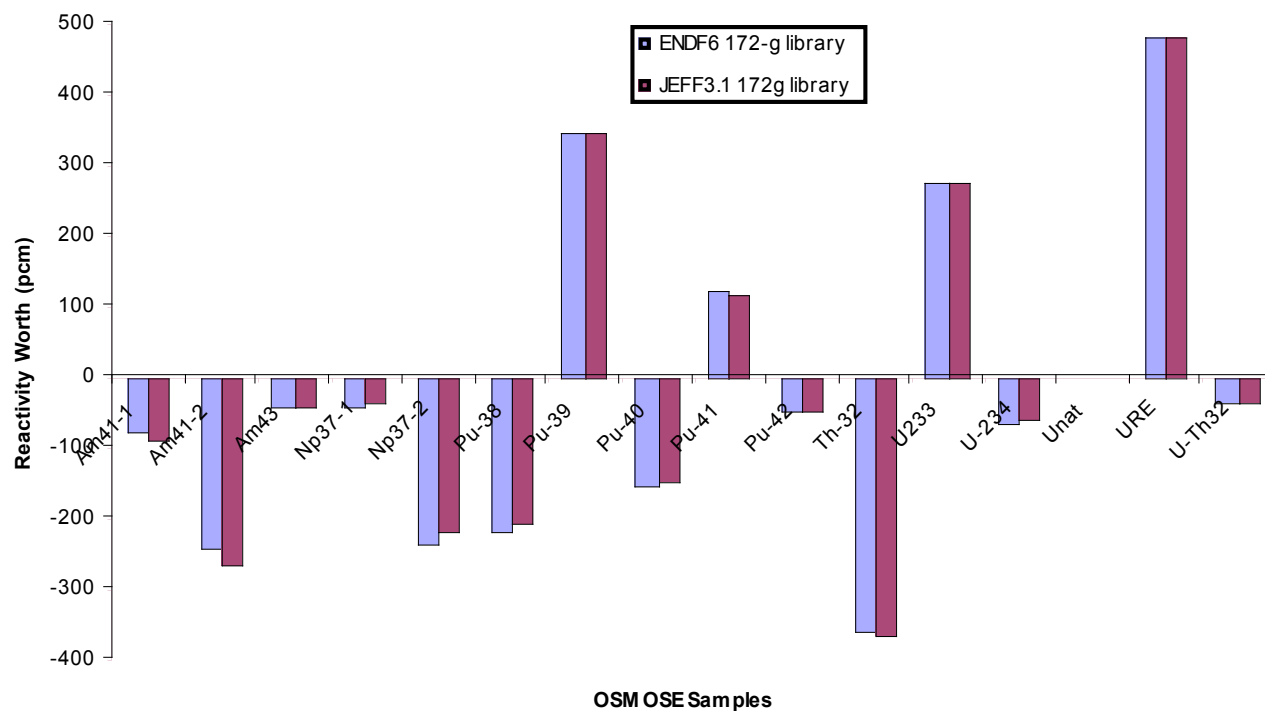
worth of the samples is referenced to the natural uranium sample which has a reactivity-worth of zero. If a sample has a negative reactivity-worth then it has a higher absorption rate than natural uranium. A positive reactivity-worth implies that a sample has a lower absorption rate, however, it usually means that the sample has a higher fission rate than natural uranium.

In the DRAGON calculations, the effective multiplication factor ( $k_{eff}$ ) is calculated with a superimposed critical buckling. For the calculation of the R2-UO<sub>2</sub>, MORGANE/R and R1-MOX configurations, the critical buckling is searched to force the  $k_{eff}$  to be 1.0 for the lattice loaded with H5 oscillation sample (2.0 % enrichment of U235).

From the results, it is observed that the OSMOSE samples in the R2-UO<sub>2</sub> configuration have the largest relative reactivity-worth, and the MORGANE/R configuration have the smallest reactivity-worth. This trend is as expected and is due to the neutron energy spectra in the sample location. As the spectra becomes harder, i.e. the thermal fraction decreases and the fast fraction increases, the number of neutron interactions in the sample decreases due to the lower thermal neutron flux. The relative reactivity-worth is subsequently reduced because it is a measure of the reaction rates (fission and absorption) in the sample.

**Table 1: Reactivity-Worth of OSMOSE Samples in the R2-UO<sub>2</sub> configuration**

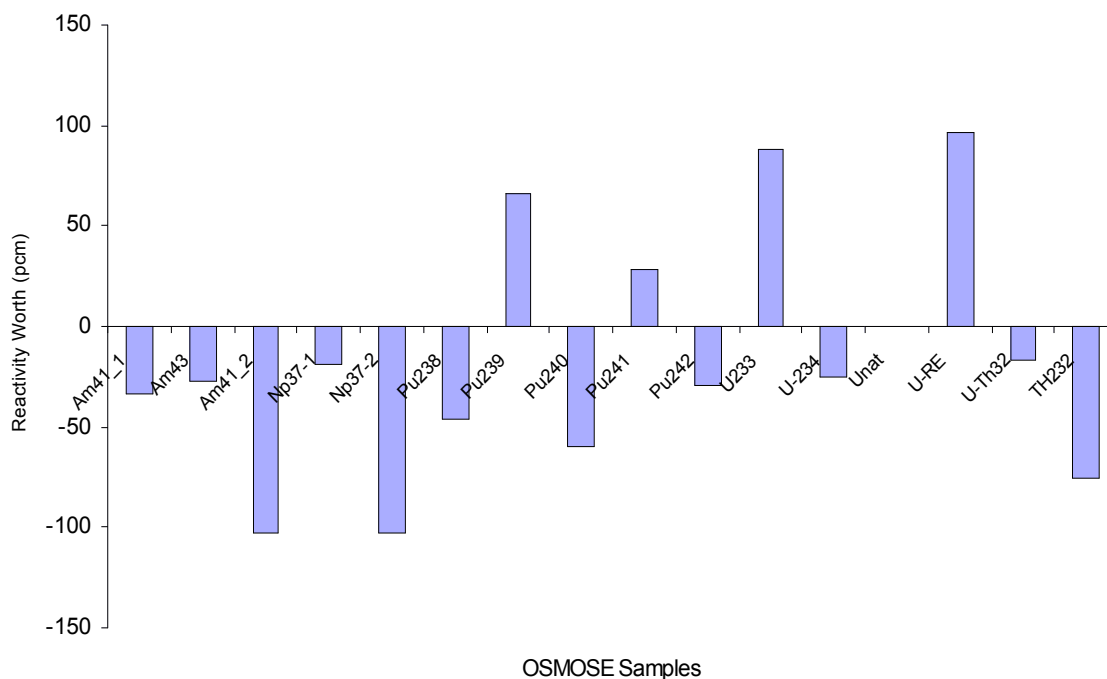
Samples	ENDF6 172-group library		JEFF3.1 172-group library	
	k-eff	reactivity worth (pcm)	k-eff	reactivity worth (pcm)
AM41_1	0.99607	-78.3	0.99595	-86.8
AM41_2	0.99444	-243.0	0.99416	-267.7
AM43	0.99642	-42.9	0.99640	-42.0
NP37_1	0.99643	-41.6	0.99643	-38.6
NP37_2	0.99448	-238.3	0.99463	-220.4
PU38	0.99469	-217.5	0.99475	-207.7
PU39	1.00030	346.9	1.00026	345.4
PU40	0.99533	-153.0	0.99533	-149.9
PU41	0.99806	122.1	0.99799	118.1
PU42	0.99638	-47.1	0.99636	-46.0
U233	0.99957	273.4	0.99952	272.1
U234	0.99619	-65.6	0.99620	-61.7
Unat	0.99685	0.0	0.99681	0.0
URE	1.00165	480.7	1.00160	479.0
U-TH232	0.99651	-33.9	0.99647	-34.3
TH232	0.99327	-361.1	0.99317	-367.8



**Figure 6: Reactivity-worth of OSMOSE samples in the R2-UO2 configuration**

**Table 2: Reactivity-worth of OSMOSE samples in the R1-MOX configuration**

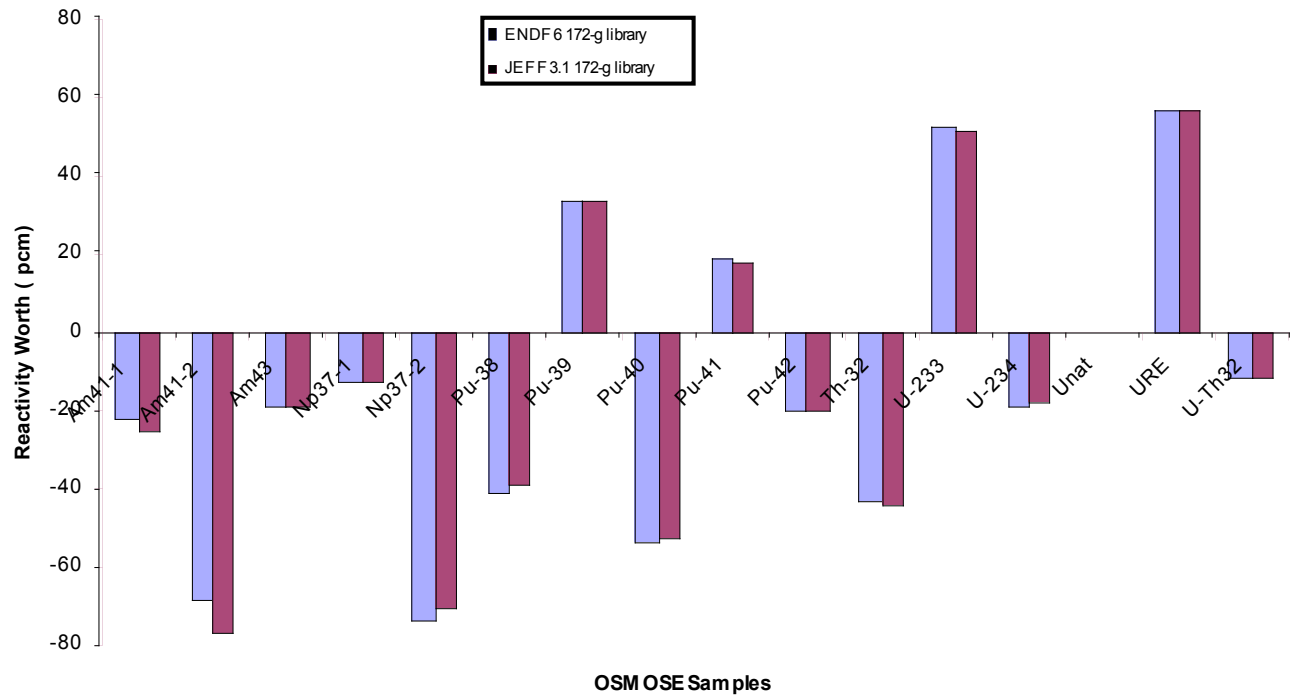
Samples	JEFF3.1 172-group library	
	k-eff	Reactivity worth (pcm)
AM41_1	0.99920	-33.84
AM41_2	0.99851	-102.70
AM43	0.99927	-27.43
NP37_1	0.99936	-18.42
NP37_2	0.99851	-103.30
PU38	0.99908	-45.66
PU39	1.00020	65.72
PU40	0.99894	-60.19
PU41	0.99982	28.02
PU42	0.99925	-29.14
U233	1.00042	87.60
U234	0.99929	-24.73
Unat	0.99954	0.00
URE	1.00051	96.70
U-TH232	0.99937	-17.22
TH232	0.99878	-122.15



**Figure 7: Reactivity-worth of the OSMOSE samples in the R1-MOX configuration**

**Table 3: Reactivity-worth of the OSMOSE samples in the MORGANE/R configuration**

Samples	ENDF6 172-group library		JEFF3.1 172-group library	
	k-eff	reactivity worth (pcm)	k-eff	reactivity worth (pcm)
AM41_1	0.99917	-22.2	0.99912	-25.3
AM41_2	0.99872	-68.1	0.99861	-76.8
AM43	0.99921	-18.7	0.99919	-18.5
NP37_1	0.99927	-13.0	0.99925	-12.5
NP37_2	0.99866	-73.5	0.99867	-70.7
PU38	0.99900	-40.8	0.99899	-39.1
PU39	0.99736	34.0	0.99972	34.0
PU40	0.99886	-53.4	0.99885	-52.4
PU41	0.99958	18.6	0.99955	17.7
PU42	0.99919	-20.2	0.99918	-19.8
U233	0.99992	51.9	0.99989	51.6
U234	0.99921	-18.7	0.99920	-17.4
Unat	0.99940	0.0	0.99938	0.0
URE	0.99997	57.0	0.99994	56.7
U-TH232	0.99928	-11.2	0.99926	-11.4
TH232	0.99897	-42.6	0.99893	-44.6



**Figure 8: Reactivity-worth of the OSMOSE samples in the MORGANE/R configuration**

### Validation and C/E Comparison

To validate the calculations, a comparison was performed between experimental measurements and calculated results for the R1-MOX configuration loaded with calibration and oscillation samples. The calculation model is based on the DRAGON lattice physics code using the IAEA 172-group JEFF3.1 cross-section library.

The process for comparison can be summarized as following:

- 1) Perform measurements, to obtain the experimental signal for calibration samples with well-known cross sections and OSMOSE samples
- 2) Calculate the  $k_{eff}$  for the calibration samples with well known cross section using the validated model, compare it to the experimental signal obtained from step 1) to generate the calibration curve ( linear function).
- 3) Calculate the  $k_{eff}$  for the OSMOSE samples using the validated model, compare it to the calibration curve obtained from step 2), if there exist significant difference, it is generally due to the cross section.

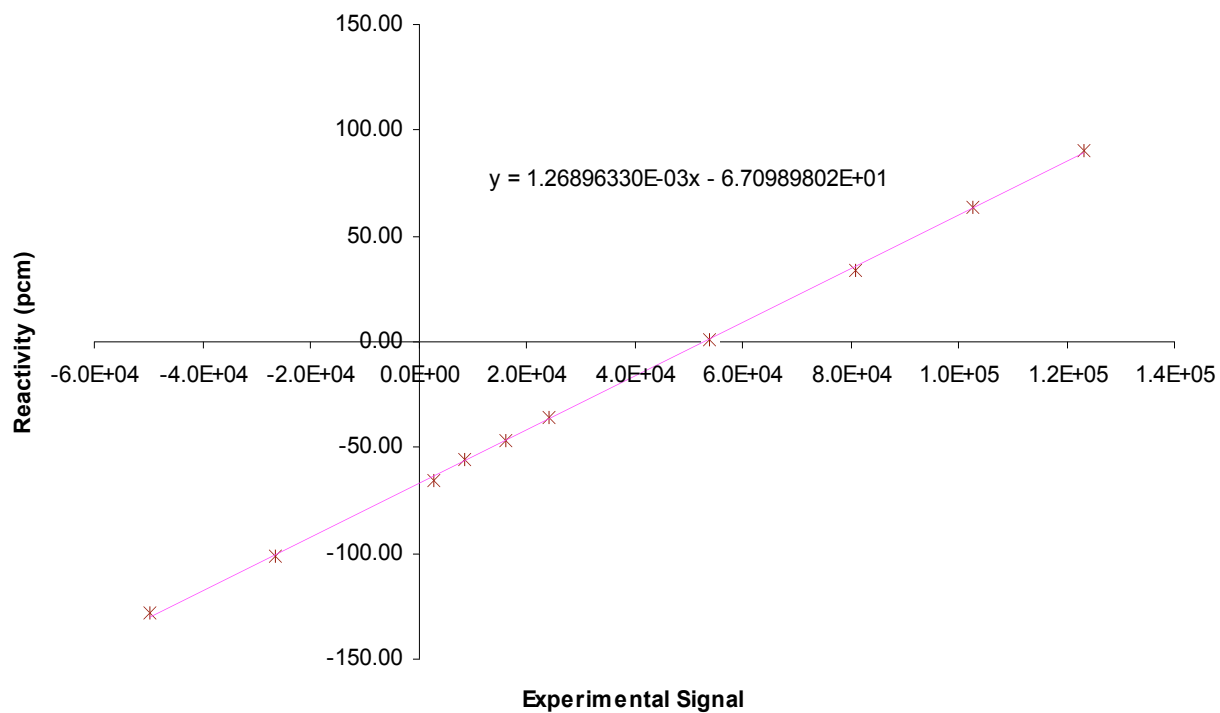
For the R1-MOX configuration, the comparison of calculated results and experimental data has been performed using preliminary experimental results. Table 4 shows the calculated  $k_{eff}$  of the B-10 and U-235 calibration samples, as well as the experimental signal (in pilot units).

**Table 4: Calculated eigenvalue and experimental signal of the calibration samples**

Sample	Enrichment of U235 (wt. %)	Boron Density (ppm)	$k_{eff}$	Experimental Signal
H1	0.25	0	0.99935	2,834
H2	0.50	0	0.99945	8,711
H3	0.71	0	0.99953	16,300
H4	1.00	0	0.99964	24,001
H5	2.00	0	1.00001	53,997
H6	3.00	0	1.00034	80,958
H7	4.00	0	1.00064	102,595
H8	5.00	0	1.00091	123,183
1B0150	0.25	150	0.99899	-26,381
2B0333	0.53	333	0.99872	-49,717

Figure 9 is the calibration curve, which shows the relation between the experimental signal (in pilot units) of the B-10 and U-235 calibration samples and the calculated eigenvalues (given by the DRAGON 2D model). It has been observed that for the calibration samples, ( the composition is UO<sub>2</sub> fuel with different enrichments in <sup>235</sup>U and with a range of boron concentrations ), the calculated reactivity worth is almost perfectly linear with the value of experimental signal, as shown in Figure 9, with RMS less than 0.02. This shows that the data for the major actinides - <sup>235</sup>U and <sup>238</sup>U, in the ANL ENDF/B-VI library is sufficiently accurate over this energy region.

Using the function between  $k_{eff}$  and experimental signal determined in Figure 9, as well as the corresponding experimental signals for the OSMOSE samples in the R1-MOX configuration, the “experimental” reactivity-worth and calculated reactivity-worth of the OSMOSE samples can be predicted. The comparison to the calculated results is shown in Table 5. It can be observed that the agreement between the calculated and experimental results is excellent for some of the samples and marginally acceptable for most of the other samples. However, for the Pu242 sample there is a large discrepancy between the calculated and experimental values. The results are being reviewed. These comparisons are only preliminary, because the experimental signals have not been validated yet and the sample analysis results are still pending. Each of these factors could easily explain the discrepancy in the results for the Pu242 sample as well as others.



**Figure 9: Calibration curve for R1-MOX configuration**

**Table 5: C/E Comparison of the OSMOSE samples in the R1-MOX configuration**

OSMOSE Samples	Calculated Reactivity-Worth (pcm)	Experimental Reactivity-Worth (pcm)	(C-E)/E in %
Np237-1	-18.42	-17.33	6.31
Np237-2	-103.30	-97.57	5.88
Pu239	65.72	64.46	1.95
Pu242	-29.14	-24.20	20.39
U234	-24.73	-22.93	7.84
U-Th232	-17.22	-17.23	-0.05
U-RE	96.70	97.98	-1.32
Am41-1	-33.84	-34.16	-0.93
Am41-2	-102.70	-96.31	6.63

## Conclusions

Initial estimates of the reactivity worth of the OSMOSE samples for the have been calculated for the R2-UO<sub>2</sub> and MORGANE/R configurations.

Since the neutron spectrum in the R2-UO<sub>2</sub> configuration is considered to be over-moderated, the results are consistent with analytic results and what one would expect for a thermal spectrum. The samples containing isotopes of fissile materials with a fission cross-section at thermal energies have a reactivity worth larger than the natural uranium sample. Isotopes that do not fission at thermal energies have a lower reactivity worth than the natural uranium sample.

From the results, it is also observed that the R2-UO<sub>2</sub> configuration loaded with OSMOSE samples has the largest reactivity worth, whereas the MORGANE/R configuration loaded with OSMOSE samples has the smallest reactivity worth. This is because the R2-UO<sub>2</sub> configuration has the softest spectra which corresponds to the largest thermal flux, while the MORGANE/R configuration has the hardest spectra which corresponds to the smallest thermal flux.

Preliminary comparison between the calculated and experimental results has been performed for the R1-MOX configuration. The C/E agreement is fairly good in general, but still needs to be studied once the experimental measurement results and the sample compositions have been confirmed. Work will continue to validate the experimental signals and sample compositions.



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